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Applicant : NG et al                      Examiner : Vanessa T VELASQUEZ  
Series No. : 10/542721                      Group Art Unit : 1793  
Filed : September 26, 2005                      Docket : 102881-11  
Title : AN IMPROVED SMELTING PROCESS FOR THE PRODUCTION OF IRON

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DECLARATION

1. I, Dr. A. Markus Reuter, of Apartment 6, 284 Williams Road, Toorak, Victoria 3142, Australia, am an internationally recognised expert in metallurgy and resource engineering. I have extensive academic and industry metallurgical experience, gained in several countries including South Africa, Germany, The Netherlands and Australia, primarily in non-ferrous metallurgy (including copper recovery), but also including ferrous metallurgy. I have a B.Eng. Degree (Honours, Cum Laude) in Chemical/Material Process Engineering, an M.Eng. Degree in Material Process Engineering and Ph.D. in Extractive Metallurgy, all from the University of Stellenbosch, South Africa. I have a Dr. habilitation from Aachen University of Technology, Germany and a D.Eng. (Doctorate in Engineering), also from the University of Stellenbosch. I was a Professor of Sustainable Technology, and now am a Professorial Fellow of Melbourne University, Australia, and I was Extraordinary Professor of University of Stellenbosch. I am Professor Emeritus, Delft University of Technology, the Netherlands.

2. I am a Registered Professional Engineer in South Africa, and a Fellow of the Australian Institute of Engineers. I have presented numerous keynote and invited lectures and supervised many Ph.D. and M.Sc. Projects, while I have authored one technical book and contributed chapters to others, I have had over 350 papers published in refereed journals and conference proceedings and I have participated in many industrial/consulting projects. I have extensive knowledge and experience in relation to the commercial processes for smelting iron-containing source materials, including iron oxide materials. That includes knowledge and experience of top submerged lance smelting of iron-containing

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source materials, for the recovery of iron by reduction smelting of iron oxide materials such as disclosed in US patent 5498277 (the Floyd patent) and US patent application 10/542721 (the present application). A copy of my full CV is attached.

3. I have been requested to provide this declaration by an Australian Patent Attorney acting for Ausmelt Limited, the assignee for US application 10/542721. I hold the position of Chief Executive Technologist at that company. For that purpose, I have been provided with:

- (a) a copy of US application 10/542721 (with the patent claims 1 to 21);
- (b) a copy of a US patent Examiner's Office Action on application 10/542721, indicated as mailed on February 02, 2009; and
- (c) a copy of prior patents comprising:
  - (i) US5498277 to Floyd et al, (the Floyd patent) assigned to Ausmelt Limited; and;
  - (ii) US4968661 to Teller et al, (the Teller patent) assigned to The Standard Oil Company.

4. In the following, my comments are directed to the two main paragraphs at page 3 of the Office Action. My comments follow those paragraphs point by point or sentence by sentence, in order.

5. The process of claim 1 of the present application can not work with excess oxygen. Also, claim 1 of the present application in fact excludes excess oxygen. I understand that "comprising" in claim 1 of the present application allows for the possibility of other steps. However, claim 1 of the present application specifies not only steps to be performed, but conditions to be achieved by those steps. It would be inconsistent with the express requirements of claim 1 of the present application to allow for another step which negated one or more of these conditions. The top submerged injection of oxygen and fuel/reductant results in at least one reducing region in the bath while, due to this and the additional reductant added by step (b) of claim 1, the overall bath will be reducing. Excess oxygen would negate this and render the process unworkable. Also, it is to be noted that the process of claim 1 of the present application is for smelting source material having its iron present as oxide and, as specified in step (b) of claim 1 of the present application, the source material is to be subjected to smelting reduction. Excess oxygen also would prevent this.

6. It is correct that the degree of combustion will vary depending on the amount of oxygen supplied. However, given the requirements for at least one reducing region to result from step (a) of claim 1, the additional reductant of step (b) and resultant reduction smelting, the amount of oxygen supplied can not be allowed to provide excess oxygen if the process of claim 1 of the present application is to be possible. Excess oxygen will only produce iron oxide and can not produce metallic iron.

7. It is correct that the Floyd patent shows variation of the degree of combustion of from about 40% to 50%. However, it is not consistent with the Floyd patent to decide on a degree of combustion in that range and then expect to achieve that degree while providing excess oxygen. The clear requirement of the Floyd patent is that if, for example, the degree of combustion is to be 45%, the remaining 55% of the fuel/reductant, in combination with the additional reductant of step (b), is required to provide the strength of reducing conditions necessary to achieve the smelting reduction of the iron oxide source material. Oxygen in excess of that required for the 45% degree of combustion will result in a higher degree of combustion of the fuel/reductant as fuel, and a reduced level of the fuel/reductant available as reductant for the smelting reduction. Of course, at the higher degree of combustion, there still will not be excess oxygen, if there is to be viable smelting reduction to produce iron. For the smelting reduction, the total amount of reductant in the system (after allowance for reductant combusted as fuel) must be large enough to sufficiently remove all oxygen from iron oxide of the iron-containing source material. Excess oxygen would be possible only if all of the fuel/reductant and additional reductant is combusted, leaving no reductant to provide reducing conditions necessary for smelting reduction of iron oxide. In specifying a degree of combustion of the fuel/reductant of from 40% to 50%, the Floyd patent is specifying that from 50% to 60% of the fuel/reductant is to remain for involvement in the smelting reduction, a situation not possible with excess oxygen.

8. The Floyd patent specifies a degree of combustion from about 40% to about 50%. There is a significant gap between this upper level of about 50% and the lower limit of in excess of 60% in claim 1 of the present application. Also, there are very good reasons why the express teaching of the Floyd patent discloses an upper limit of 50% for the degree of combustion, and I will return to those reasons later herein. However, it is worth noting at this stage that, rather than excess oxygen, there must be excess reductant (relative to the quantity of injected oxygen) and the excess reductant will result in generation of combustion gases comprising CO and H<sub>2</sub>, as specified in step (b), necessary to enable the post-combustion required by step (d) of claim 1. That is, the degree of combustion required by the Floyd patent and the process of claim 1, results in less than complete or substantially

complete combustion, as the CO and H<sub>2</sub> are not combusted in the bath, and enable the post-combustion of step (d) above the bath.

9. It is agreed that the Teller patent indicates that complete or substantially complete combustion of carbon containing material can necessitate oxygen in excess of stoichiometric requirements. The Office Action states "must be added in excess" (underlining added), but this is a departure from the actual wording of the Teller patent in the passage from column 1, line 63 to column 2, line 5. Indeed, the only use of "must" in such a context in the Teller patent is at column 10, lines 59 to column 11, line 18, in which the reference is to a mole ratio of oxygen to methane which is sub-stoichiometric (i.e. the opposite of excess oxygen).

10. The reference in the Teller patent to complete or substantially complete combustion stands in stark contrast to the requirements of the process of claim 1 for conditions for the production of metallic iron. The reference provides no guidance in relation to a skilled person in the art of smelting reduction of oxidic iron source material. In the passage in the Teller patent referred to in the Office Action, the concern is with full combustion of a carbonaceous material to carbon dioxide and water, leaving no combustible intermediate product. That is, intermediate products such as carbon monoxide also are to be combusted. This is not possible in a smelting operation, particularly one which, like the process of claim 1, is conducted while continuing to add source material and oxygen. As explained later herein, the combustion of the carbonaceous fuel of the fuel/reductant and also the actual smelting reduction generate both carbon monoxide and hydrogen as intermediate products, and this enables the post-combustion step required by claim 1 of the present application. That is, carbon monoxide and hydrogen are produced, and they are not fully combusted to carbon dioxide and water by injected oxygen, even though the required degree of combustion of carbonaceous material has been achieved. If in the process of the Floyd patent a degree of combustion of 45% is required in step (a), then 45% of the fuel/reductant is able to be combusted, albeit with generation of carbon monoxide and hydrogen, leaving 55% of the fuel/reductant available to provide, in combination with the additional reductant of step (b), the required smelting reduction. The same applies with the process of claim 1 of the present application if, for example, 75% of the fuel/reductant is combusted in step (a), leaving 25% of the fuel/reductant as reductant, bearing in mind that some fuel/reductant is necessary, and must remain, to provide, in combination with the additional reductant of step (b), the reduction smelting. As understood by persons skilled in the art, the term "degree of combustion" of fuel/reductant in a slag phase of a bath in which the fuel/reductant also is to give rise to smelting reduction is incompatible with a concept of complete or substantially

complete combustion as discussed in the Teller patent. This is because, in that context of the smelting reduction:

- (a) there can not be excess oxygen in the slag as it would preclude reduction of iron oxide of the source material to metallic iron, and
- (b) the combustion will result in evolution of the combustible intermediate products of carbon monoxide and hydrogen.

That is, the context in which the Teller patent refers to complete or substantially complete combustion can not be related to the context of claim 1 of the present application in which a degree of combustion of 100% is precluded and is completely untenable.

11. There are numerous factors bearing on the reduction smelting processes of the Floyd patent and claim 1 of the present application to which the disclosure on combustion in the Teller patent has no bearing. These include:

- (a) combustion in the reduction smelting processes, which occurs at the tip of the lance, is conducted within a molten slag phase in which iron oxide is dispersed to be melted and taken into solution;
- (b) despite the combustion at the lance tip, net reducing conditions have to prevail in the molten slag phase in order to achieve reduction by which iron oxide is reduced to molten iron metal;
- (c) the reduction smelting to produce a molten iron metal phase necessitates a high temperature – indicated in the present application as being from 1350°C to 1500°C – but, with increasing temperature above about 800°C, combustion of carbon increasingly favours the production of carbon monoxide rather than carbon dioxide, essentially precluding complete or substantially complete combustion as in the Teller patent, even if this was not already precluded by the need for iron oxide reduction.

These factors are parts of the basic knowledge of persons skilled in the art of those reduction smelting processes. That basic knowledge leads to a necessary recognition that complete or substantially complete combustion as disclosed in the Teller patent is not remotely possible in those processes.

12. I have mentioned in paragraph 8 that I would return to the good reasons why the Floyd patent expressly sets an upper limit of a degree of combustion of 50%. The implication of the relevance of the Teller patent to the present invention, as asserted in the Office Action, is that fuel/reductant can be fully combusted. Thus, if the fuel/reductant in claim 1 of the present application is carbon, the assertion is that it will be combusted by the reaction:



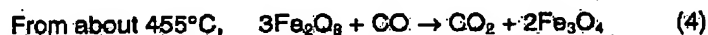
Even if this was the case, the wording of claim 1 would preclude excess oxygen due to the requirement that sufficient un-combusted carbon is to remain available to act as a reductant producing metallic iron. However, as intimated in the last paragraph, reaction (1) is not favoured above 800°C, and certainly not at temperatures from 1350°C to 1500°C able to be used in the process of claim 1 of the present application. Rather, the reaction:



is favoured, while  $\text{CO}_2$  produced can undergo the reaction:



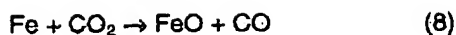
Also, the reduction reactions involving iron oxide to be smelted are complex, as follows:



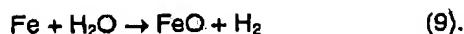
or



Reactions (1) to (3) occur simultaneously with reactions (4) to (7). In addition, water vapour usually is present from the carbon (coal) and combustion of hydrogen. Carbon dioxide (contrary to any suggestion based on the Teller patent) and also water vapour are undesirable as they can give rise to reversal of reaction (6), producing a lower recovery of metallic iron due to iron oxide (FeO) being lost in the slag, i.e.:



as well as



That is, each of reactions (8) and (9) reverses production of metallic iron.

13. At typical temperatures for the process of the Floyd patent, thermodynamic data indicate a need for a very low oxygen potential. The attached Ellingham Diagram is a plot of Gibbs free energy against temperature for metals reacting to form oxides at an oxygen partial pressure of one atmosphere, and enables a determination of the oxygen potential or partial pressure in equilibrium with the metal and oxide at a given temperature. At a selected temperature, say 1400°C, a line from zero on the Gibbs free energy scale at the left side of the diagram, is drawn through the intersection of the vertical line for 1400°C with the line for, say, the reaction:



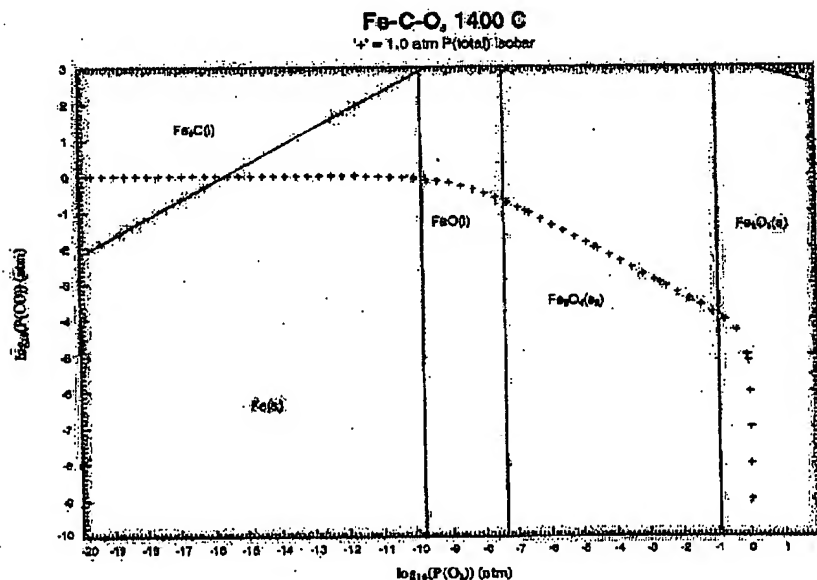
That line, when continued to intersect the oxygen partial pressure scale on the right hand side of the diagram, gives the oxygen partial pressure for reactions (6) and (7), as well as the CO/CO<sub>2</sub> ratio of around 5/1. That line indicates an oxygen potential of about 10<sup>-10</sup> atmospheres is required to sufficiently reduce iron oxide to metallic iron. That is, thermodynamic data for the production of iron by smelting reduction processes for oxidic iron materials, including that of the Floyd patent, indicates that such low levels of oxygen potential need to prevail. In practice, the oxygen potential is lower than 10<sup>-10</sup>, since the iron oxide is to be reduced in the slag phase in which the activity of FeO is lower than 1, and an oxygen potential of lower than 10<sup>-12</sup> atmospheres is required. Moreover, industrial practice with these processes is consistent with this thermodynamic data, no doubt because those persons skilled in the art are familiar with and rely on such data. It is on such thermodynamic data and practical experience that the upper limit of 50% degree of combustion in the Floyd patent is based. The thermodynamic data and industrial practice were thought to fit well with other factors. The degree of combustion of 40% to 50% was considered to achieve what was perceived as necessarily strong reducing conditions for optimum metallic iron recovery, while limiting the combustion of fuel/reduction to a level sufficient to provide adequate temperatures of 1350°C to 1500°C. At such operating temperatures the molten iron was to be sufficiently molten to have a content of about 3% to 4% of carbon in solution, and the upper limit for the degree of combustion of 50% was believed appropriate for suppressing

reactions (8) and (9). Thus, there were compelling reasons why the Floyd patent specifies a degree of combustion of, in effect:

- (i) not less than 40%, and
- (ii) not more than 50%.

That is, operating in the range of 40% to 50% is an express requirement of the Floyd patent.

14. In this paragraph there is shown a plot for the Fe-C-O system of the log of the partial pressure of carbon monoxide against the log of the partial pressure of oxygen, at a temperature of 1400°C. In this, points shown by the symbol "+" are on the 1.0 atmosphere isobar for total pressure. A person skilled in the art applicable to the process of the Floyd patent would appreciate from this plot, as well as basic requirements for reduction smelting, that the combustion of fuel/reductant and additional reductant must never be complete in order to ensure that metallic iron is produced.





15. Based on my comments in paragraphs 5 to 14, I consider that it clearly is incorrect to suggest that "to obtain a degree of combustion between 50%, as taught in Floyd and 100% (completeness), as taught by Teller, it would have been obvious to one of ordinary skill in the art to optimise the amount of oxygen in the system", as asserted in the first paragraph, page 3 of the Office Action. That assertion is technically unsound. It is contrary to what the Floyd patent makes clear is an upper limit for oxygen, i.e. an amount of oxygen providing a degree of combustion of only from 40% to 50%. A person skilled in the art would not be aware of any motivation to depart from the Floyd patent. Such a person would recognise that to depart from the Floyd patent was contrary to available data and practice, and that complete or substantially complete combustion of the fuel/reductant would result in little if any recovery of metallic iron. The skilled person also would appreciate that there is no issue of optimising the amount of oxygen in the system. To elaborate on these issues, it should be noted that at column 2, lines 11 to 27 the Teller patent points out the extent of excess oxygen required for various fuel types, ranging from usage of air (and hence oxygen) 10 to 50%, and more, in excess of stoichiometric requirements, in order to achieve complete or substantially complete combustion. These excesses are said to have disadvantages. To suggest that it would have been obvious "to obtain a degree of combustion between 50%, as taught in Floyd, and 100% (completeness), as taught by Teller" thus ignores the fact that the Floyd patent in effect, states not more than 50% and that the Teller patent, in effect, achieves completeness (if this means a degree of combustion of 100%) by having a 10% to 50% excess of oxygen beyond stoichiometric requirements.

16. In the second paragraph of page 3, the Office Action comments, that "the portion relied on in Teller would be common general knowledge to one of ordinary skill who works in a field requiring an understanding of combustion (e.g. smelting)". Except for the reference to smelting, this no doubt is correct. However, that portion of the Teller patent has no relevance to a context in which a region of combustion is required within an environment in which reducing conditions are to prevail. The combustion required in the process of claim 1 of the present application, which is limited to a region at the lance tip, is to provide heat to attain a temperature, such as 1350°C to 1500°C, at which the slag is molten, the iron oxide source material can be melted by the slag and taken into solution, molten metallic iron product is at a suitable temperature, and the required smelting reduction reactions are able to proceed to produce the iron product, with all of this being achieved within a slag phase. The wording "understanding of combustion (e.g. smelting)" used in that second paragraph, page 3 of the Office Action, is completely inapt. Combustion involves oxidation. However, while there are oxidative smelting processes such as for roasting sulphides, the invention of claim 1 of the present application requires smelting under reducing conditions. The issue clearly is not one

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